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INVESTIGATING THE STATE OF A 6 YEARS OLD FULL-SCALE BIOCOVER AT A DANISH LANDFILL

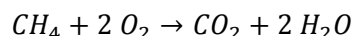
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SUMMARY: A biocover system was established at Klintholm landfill in Denmark in 2009 to mitigate methane emissions originating from landfill gas escaping due to advection and diffusion processes. The system consisted of nine sections of crushed concrete, which acted as a gas distribution layer, beneath a layer of compost in an otherwise low permeable top cover. The purpose of the compost layer was mainly to provide suitable living conditions for bacteria able to oxidize methane into carbon dioxide. A series of field and laboratory tests was carried out to evaluate the current performance of the biocover system. Field tests showed total methane emissions on the same level or lower than previous measurements and laboratory tests showed methane oxidation potentials approximately equal to former tests. In spite of an inhomogeneous distribution of landfill gas to the methane oxidation layer, it was concluded that the performance of the biocover system had not declined over the 6-7 years since its establishment, even though no maintenance had been carried out in the intervening years.

1. INTRODUCTION

The potent greenhouse gas methane (CH₄) is generated in landfills due to decomposition of organic waste. If not collected and treated, CH₄ may emit to the atmosphere and contribute to global warming. Biocover systems have been proposed as a mitigation action in situations where gas collection and utilization is not economically or technically feasible. The idea of a biocover system is to lead generated landfill gas through a gas distribution layer (GDL) and on to a filter material containing methanotrophic bacteria. These bacteria are able to assimilate CH₄ or oxidize it into carbon dioxide (CO₂) as follows:



As, in this case, the CH₄ originates from organic waste, the generated CO₂ is biogenic, which is assigned a global warming potential of zero.

Due to the novelty of the technology, there is limited knowledge about the long-term efficiency and maintenance needs of full-scale biocover systems. These parameters are important when assessing the overall cost of a biocover system. Previously, the efficiency of a 6 years old experimental biocover at St-Nicéphore landfill in Quebec, Canada has been investigated (Capanema et al., 2013). They found an increase in vegetation on the biocover surface and demonstrated CH₄ oxidation efficiencies of 73-100%. In this paper, the functionality of a 6 years old full-scale biocover system has been evaluated. This biocover was subject to research at its establishment and thus monitored and tested thoroughly. Many of those findings are presented in (Scheutz et al., 2014). In 2016 and 2017, the site was revisited to investigate whether the biocover still functioned as intended.

It has previously been shown that landfill gas emissions and soil gas compositions are dynamic and influenced by factors such as barometric pressure and temperature (Christophersen et al., 2001; Czepiel et al., 1996; Gebert et al., 2011; Gebert and Gröngroft, 2006). This has to be taken into account when analysing data and comparing findings. Too high or low temperatures and water content in the methane oxidation layer (MOL) affect the bacterial activity negatively (Scheutz et al., 2009). In Denmark, the monthly average temperature ranges from 0-16 °C and the monthly precipitation ranges from 38-79 mm (Danmarks Meteorologiske Institut, 2017). Therefore, periods of desiccation are unlikely while inhibition due to low temperatures may occur. Some of the results presented in this paper are preliminary, as research is still being carried out at the site.

2. SITE CHARACTERISATION

Klintholm landfill, located on the Danish island Funen, was established in 1978 and consists of several waste cells, some still active. The oldest cell is Cell 0, which was active from 1980-1996. It has an estimated waste volume of 485,000 m³ and contains a mix of combustible waste, sludge, and non-combustible waste. In 2009, a biocover system was established at this cell. The system consists of piping leading gas from the interior of the landfill to nine sections with a GDL consisting of crushed concrete (30 cm) situated beneath a filter material consisting of compost (70 cm). The compost originated from a mix of garden waste and kitchen waste in a 7 to 2 ratio. The surface area of the biocover is approximately 4800 m². No maintenance of the biocover had been carried out in the years between its establishment and the time this paper was written.

3. METHODS

A visual inspection of the biocover was made to investigate the state of the biocover surface. The total CH₄ emission from the landfill was estimated using a tracer dispersion method utilizing a Cavity Ring Down Spectroscopy based system (Picarro Inc. USA). The method is described in (Mønster et al., 2014) and the implementation of this method at Klintholm landfill is further described in (Scheutz et al., 2014). Near surface concentrations of CH₄ was measured using a hand-held Laser One (Huberg S.a.s., Italy) CH₄ detector at the entire biocover surface. 16 measuring points in a 4 X 4 grid was selected in three of the biocover sections. In each of the 48 points, surface emission rates of CO₂ and CH₄ were measured using a flux chamber connected to an Innova 1312 photoacoustic multi gas monitor (LumaSense Technologies A/S, Denmark). A more detailed description of the method and

materials used for flux chamber measurements is given in (Scheutz et al., 2011). In the same points, gas samples were recovered from the filter material using probes and analysed on site for CO₂, CH₄, and O₂ using a BIOGAS 5000 Gas Analyser (Geotechnical Instruments Ltd., United Kingdom). Gas samples were obtained for every 10 cm by pushing a steel probe (inner diameter of 6 mm, closed in the bottom and slotted over the bottom 5 cm) vertically through the filter material. When reaching the GDL in each point, it was not possible to push the gas probes further by hand and the final gas sample was recovered. The temperature was measured using a thermometer in each of the measuring points. As for the gas sampling, the temperature was measured throughout the filter material for every 10 cm. The thickness of the filter material was measured in the 48 points using the gas sampling probes and the thermometer as indicators. For an overview of Cell 0, see Figure 1.

Three measurement campaigns were carried out to capture different barometric pressures and low atmospheric temperatures at which the working conditions for the methanotrophic bacteria were assumed poor: One campaign in March 2016 during increasing barometric pressure, one campaign in April 2016 during stable barometric conditions, and one in January 2017 during a decreasing barometric pressure. All field measurements were carried out at two or more of the campaigns.

Excavations were made in each of the three selected sections in March 2016 to recover filter material for laboratory experiments. Samples of the filter material were taken for every 10 cm in a vertical line between the surface and the GDL and stored in plastic containers at 10 °C until the laboratory experiments commenced.



Figure 1. Cell 0 seen from above. The grey boxes indicate the biocover sections. Pipes can be seen on the slopes of the landfill, feeding landfill gas to the sections. Blue dots represent the approximate position of the measuring points, though the area they cover is exaggerated. Orange dots represent the approximate position of the excavation sites.

Filter material samples were tested in duplicate for water content and loss on ignition. Batch incubation tests were set up to test the CH_4 oxidation potential and the respiration rates of the filter material. For these tests, approximately 35 g of filter material from each section and depth was added to a 500 mL glass infusion bottle sealed with a butyl rubber stopper and an aluminium screw cap. Infusion bottles with no filter material were used for controls. All incubation tests were performed in duplicate. For the CH_4 oxidation potential tests, the gas concentration in each batch was adjusted from atmospheric conditions to approximately 15% (vol.) CH_4 and 35% (vol.) O_2 at 1 atm. For the respiration tests, the gas concentration was adjusted from atmospheric conditions to approximately 35% (vol.) O_2 at 1 atm. After adjustment of the gas concentrations, 0.2 mL gas samples were extracted manually by syringe every few hours. The gas samples were injected into two TRACE™ 1310 Gas Chromatographs (Thermo Fischer Scientific, Massachusetts, USA) for analysis of CO_2 , CH_4 ,

and O₂ concentrations. Plotting the gas concentrations over time yielded gas concentration curves. By fitting zero-order trend lines to the near linear part of the concentration curves, it was possible to calculate maximum CH₄ oxidation rates and respiration rates.

4. RESULTS AND DISCUSSION

4.1 Visual inspection

A visual inspection of Cell 0 revealed that weeds, grass, and small bushes and trees grew on the biocover surface. At a few places, bare spots were located, presumably expressing increased CH₄ flux. Tunnels dug by rodents were observed several places and burrows made by larger animals were found as well. We observed no discolouration associated with the presence of exopolymeric substances.

4.2 CH₄ emission from Cell 0

The main goal of implementing a biocover system on Klintholm landfill was to mitigate CH₄ emission. Measuring the CH₄ emission was therefore a priority when evaluating the performance of the biocover.

4.2.1 Total CH₄ emission based on the tracer dispersion method

In March 2016 during increasing barometric pressure, the average emission from Cell 0 was measured to be 0.59 ± 0.25 kg CH₄/hour (average of 5 measurements \pm 1 standard deviation) using the tracer dispersion method. In January 2017 during decreasing barometric pressure, the average emission from Cell 0 was measured to be 0.97 ± 0.47 kg CH₄/hour (average of 7 measurements \pm 1 standard deviation) using the tracer dispersion method. In both cases, the emission is likely to be overestimated though, as two other CH₄-emitting sources contributed to the results. Previously, the average emission from Cell 0 has been reported as 1.8 kg CH₄/hour in 2010 and 0.7 kg CH₄/hour in 2011/2012 (Scheutz et al., 2014). The 2016-2017 results indicate that the overall emissions from the landfill are smaller or at the same level as previous measurements. Therefore, they give no reason to suspect a diminished efficiency of the biocover system.

4.2.2 Near surface concentration of CH₄

A surface screening of the biocover sections was performed in March 2016 during an increasing barometric pressure. It showed CH₄ concentrations of 1-10 ppm just above the MOL. Only in one point (in the northern part of Section 2) was a substantially higher concentration measured: 50 ppm. A second screening was made in January 2017 during a decreasing barometric pressure at which elevated concentrations (>10 ppm) were found more frequently. During this screening, elevated concentrations were found once or twice in Section 1, 2, 4, 7, and 9. In Section 3, elevated concentrations were found in areas in the central and in the southern part. At three spots in the southern part of Section 3, a concentration of >100 ppm was found with a maximum of 218 ppm CH₄. These findings generally do not correspond well to the findings of the surface screenings conducted shortly after the biocover's establishment, where elevated CH₄ concentrations were observed mainly at the edges of Section 2, 4, 7, and 9 (Scheutz et al., 2014). While elevated concentrations were found

in Section 2 and 4 in 2010 as well as in 2017, the older screenings showed background level concentrations of CH_4 in the southern part of Section 3 in which the highest concentrations were measured in 2017. In 2010, the surface screenings were conducted at decreasing barometric pressure.

The surface screenings suggest that CH_4 could be observed in similar maximum concentrations in 2017 as in 2010 during decreasing barometric pressure. However, in 2017, elevated concentrations were less widespread and found mostly at single points as opposed to in 2010 where large areas were subject to elevated concentrations.

4.2.3 CH_4 surface flux measured by flux chamber

During increasing barometric pressure, a flux of CH_4 through the filter material was observed in only a few of the measuring points. Positive CH_4 fluxes were observed more frequently during stable and decreasing barometric pressure, see Figure 2. The inhomogeneity of the fluxes was pronounced. Points with CH_4 fluxes greater than $50 \text{ g/m}^2/\text{d}$ were found close to points with negative fluxes. One example was at the April 2016 campaign in Section 4, where the CH_4 flux in point 2 was $-0.06 \text{ g/m}^2/\text{d}$ while the CH_4 flux in point 7, situated a few metres away, was $126 \text{ g/m}^2/\text{d}$. The greatest CH_4 flux was observed at the April 2016 campaign in Section 2, point 1; $955 \text{ g/m}^2/\text{d}$.

Flux chamber measurements carried out in Section 2 and Section 4 in 2010 showed positive CH_4 fluxes at the edge of the biocover towards its slope while all other measurements were below detection limit (Scheutz et al., 2014). There was no such recognizable pattern at the 2016-2017 measurements though positive CH_4 fluxes were observed more frequently.

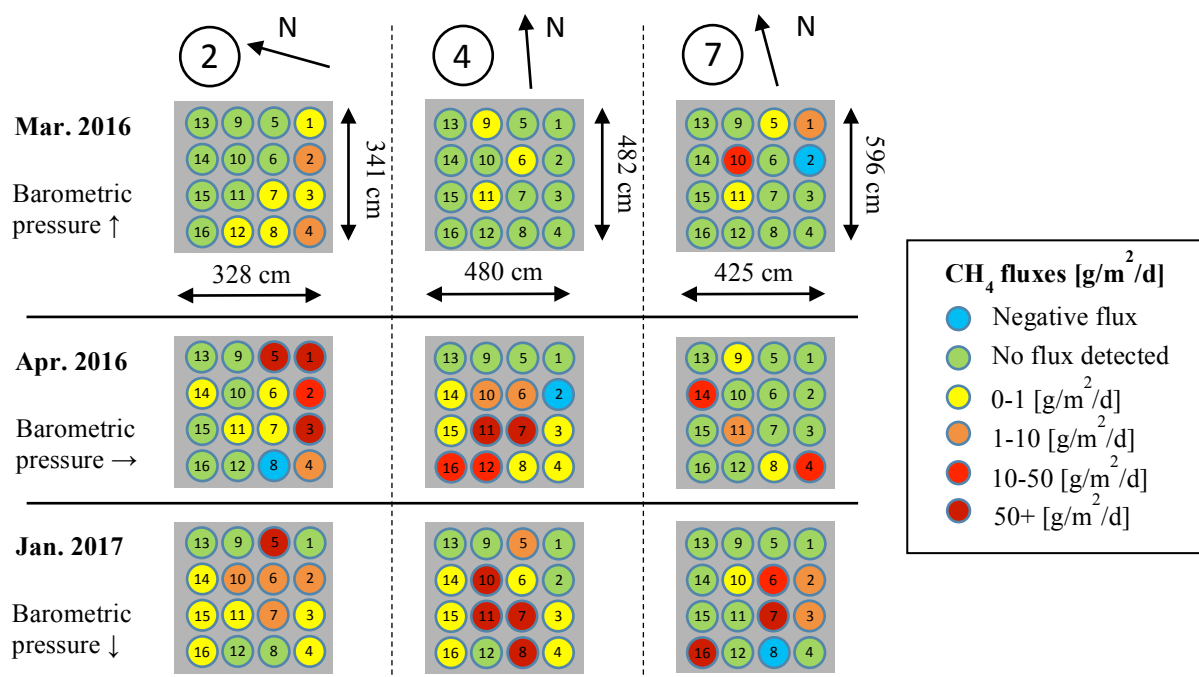


Figure 2. CH_4 fluxes through the filter material measured by static flux chamber. Upper row represents

data from March 2016, middle row represents data from April 2016, and the lower row represents data from January 2017. Leftmost column represents data from the measuring points in Section 2, middle column, the measuring points in Section 4, and rightmost column, the measuring points in Section 7. The distances in cm show the length from point no. 1 to point no. 4 and from point no. 16 to point no. 4 in each section. The measuring points were distributed in a grid, though not completely uniformly, but have been depicted so for the reader's consideration.

4.3 Gas movement and gas conversion in the filter material

When measuring gas concentrations in the MOL, CH_4 concentrations $>1\%$ was only observed in 5 of the 48 measuring points at the March 2016 campaign during increasing barometric pressure. In the majority of the measuring points, O_2 concentrations of $>10\%$ was found all through the MOL. Examples of this can be seen in Figure 3.

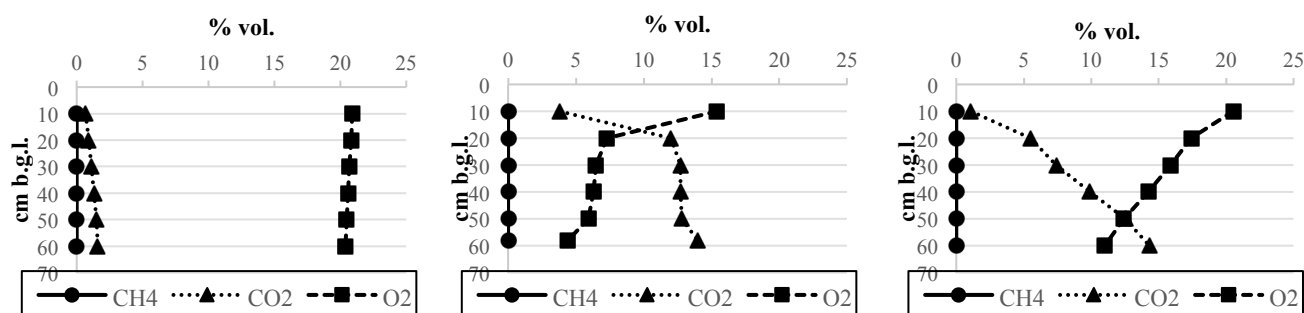


Figure 3. Gas concentration profiles of CH_4 , CO_2 , and O_2 measured in March 2016 in Section 2 point 5 (left), Section 4 point 6 (middle), and Section 7 point 11 (right). b.g.l. = below ground level.

These findings align well with the overall results of the surface screening and surface flux measurements from the same measuring campaign that indicated near atmospheric gas concentrations at the biocover surface. The gas concentration profiles further show that O_2 was able to penetrate the entire MOL. It therefore seems unlikely that the advective flux had been inhibited even though the compost that made up the MOL matured significantly from the time of the biocover's establishment to the 2016 measuring campaign.

CH_4 was observed in the MOL more frequently at the April 2016 and January 2017 campaigns. At nearly all measuring points, CH_4 concentrations were near 0% at the shallowest measured depth, though. Looking at some of the concentration profiles from April 2016, it can be seen that both CO_2 and CH_4 concentrations decrease through the MOL as it is transported from GDL below towards the surface, see Figure 4. This happens partially as the landfill gas is diluted in the MOL. However, the CO_2/CH_4 relationship increases towards the surface (until CH_4 concentrations approach 0%). This increasing relationship is an indication of the conversion of CH_4 into CO_2 . As O_2 was available in the entire MOL, CH_4 oxidation was not restricted to shallowest 40 cm, as otherwise common in landfill soil covers (Scheutz et al., 2009). Furthermore, the ratio of CH_4 to CO_2 indicated CH_4 oxidation in the GDL below the MOL.

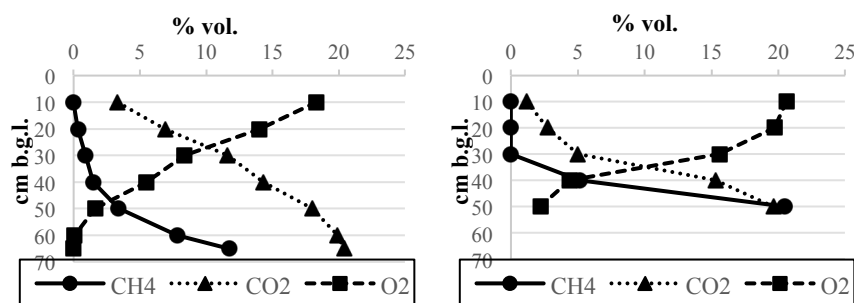


Figure 4. Gas concentration profiles of CH₄, CO₂, and O₂ measured in Section 4 measuring point 14 (left) and Section 7 measuring point 10 (right) at the campaign of April 2016.

When comparing gas concentration profiles, it became evident that the profile connected to one point was not necessarily indicative of neighbouring points. One example was the gas concentration profiles at point 7 and point 8 measured in Section 7 in January 2017, see Figure 5. Though the points were measured within a few minutes and were situated only metres apart, one profile shows no apparent CH₄ and near atmospheric levels of O₂ while the other shows an increasing CH₄ concentration towards the bottom of the MOL and an O₂ level that decreases to approximately zero. The gas concentration profiles confirmed what was observed at the surface flux measurements, namely that landfill gas is not distributed uniformly to the MOL.

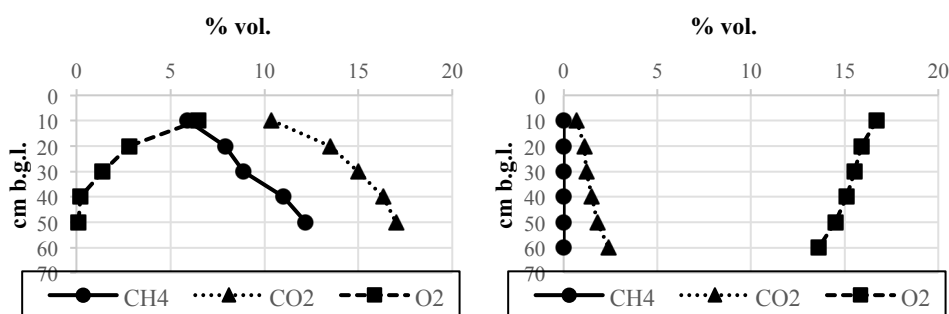


Figure 5. Gas concentration profiles of CH₄, CO₂, and O₂ measured in measuring point 7 (left) and point 8 (right) in Section 7 in January 2017.

4.4 Temperature in the filter material

Heat is provided to the MOL by the landfill gas as temperatures inside landfills can reach more than 50 °C (Hanson et al., 2010). Furthermore, heat is generated by the microbial oxidation and respiration processes. As the MOL consisted of compost and the gas concentration curves suggest CH₄ oxidation, both of these processes are expected to have contributed to the increased temperatures measured in the MOL at Klintholm landfill. Temperatures were measured at the March 2016 campaign and the January 2017 campaign.

Table 1. Ambient temperatures and temperatures in the MOL. [^]Ambient temperatures were recorded at Hans Christian Andersen Airport situated approximately 40 km from Klintholm landfill. The data was retrieved from www.wunderground.com. *Temperatures of every measured depth at every measuring point of each section was included in the calculation of average temperatures.

<i>Campaign</i>	<i>Section</i>	<i>Ambient temp.[^] (°C)</i>	<i>Min. temp. (°C)</i>	<i>Avg. temp.* (°C)</i>	<i>Max. temp. (°C)</i>
March 2016	2	6	8	13	23
	4	5	13	25	41
	7	6	10	27	39
January 2017	2	2	6	24	45
	4	2	8	22	35
	7	2	5	19	32

At all measuring points, temperatures were lowest at the shallowest part of the MOL and highest at the deepest part at the brink of the GDL. The average temperature found in the MOL was considerably higher than the ambient temperature, see Table 1. In January, during the Danish winter, the average temperature was close to the reported optimum temperature for CH₄ oxidation: 25-35 °C (Scheutz et al., 2009). As seen with the previously presented data, temperature measurements showed large variations from one measuring point to another (data not presented). At the March 2016 campaign, the lowest temperatures were measured in Section 2. When the temperature was measured at the January 2017 campaign, the highest temperatures were observed in Section 2. This indicates that activity in the biocover may not merely change within a section e.g. from one measuring point to another. An entire section that at one point is inactive does not necessarily remain so and vice versa.

4.5 Characterisation of the filter material

The depth of the MOL was 15 cm smaller on average than at the biocover's establishment, see Table 2. It must be recognized that measurements in 2016-2017 were restricted to small areas in only three of nine biocover sections. At the excavations, it became visible that the filter material no longer had the coarse structure of compost. Instead, the structure was much finer. This transformation had likely given rise to a natural settlement and compaction of the MOL. The loss on ignition was lower than at the biocover's establishment, which suggests that some of the organic carbon had been converted through microbial processes. This was likely another factor influencing the lowered MOL depth. There were clear indications that the microbial processes continued the degradation of organic material as the average respiration rate was similar to that previously measured. When looking at CH₄ oxidation rates and the water content, it seems the characteristics of the filter material did not change much. Bacteria in different areas and depths of the MOL seem to have specialized though, as evidenced by the increased spread in oxidation and respiration rates.

Table 2. Data from laboratory tests and selected field measurements. The values presented are averages, the uncertainty represent one standard deviation. Numbers in parenthesis represent the number of measurements or experiments the average and uncertainty is based on. DM = dry matter. n/a = not available. ^a (Scheutz et al., 2014). ^b Background data for (Pedersen et al., 2012).

<i>Measurement</i>	<i>Previous findings</i>	<i>This study</i>
Methane oxidation layer depth (cm)	70 ^a	55 ± 8 (237)
Oxidation rate in filter material at 22 °C (μg CH ₄ /g DM/h)	31.4 ± 12.5 ^a (2)	42.1 ± 23.2 (34)
Maximum oxidation rate in filter material (μg CH ₄ /g DM/h)	141 ± 11 at 30 °C ^b (2)	129 ± 18 at 40 °C (2)
Respiration rate in filter material at 22 °C (μg CO ₂ /g DM/h)	18.1 ± 4.4 ^a (2)	19.9 ± 13.8 (34)
Water content (per 100 g DM)	35.2 ± 0.6 ^a (n/a)	35.5 ± 7.6 (34)
Loss on ignition (per 100 g DM)	18.8 ± 0.3 ^a (n/a)	11.6 ± 3.3 (34)

5. CONCLUSIONS

Total emission measurements conducted by a tracer dispersion method showed CH₄ emissions at a lower or similar level compared to previous measurements. Gas concentration profiles showed clear signs of CH₄ oxidation in the MOL. Surface screenings and surface flux measurements revealed that areas with positive CH₄ fluxes were scattered and dynamic. This suggests that landfill gas was not distributed evenly between and throughout sections. Temperatures higher than ambient temperature were observed throughout the MOL with average temperatures between 13 and 27 °C, even when measured in the traditionally coldest month of the year. This indicates microbial activity and demonstrate the insulating effects of the filter material. The temperatures and the water content found in the MOL renders it unlikely that the methanotrophic bacterial was inhibited by these factors. In conclusion, the biocover system worked as efficiently as when it was first established in 2006. The results point to no or little need for maintenance in the first 6-7 years of a biocover system's lifetime and indicate that the biocover was not under scaled in the design phase.

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